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AEC RESEARCH AND DEVELOPMENT REPORT

LOS ALAMOS SCIENTIFIC LABORATORY

OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO

THE USE OF QUARTZ TUBES FOR SAMPLING AND CASTING PLUTONIUM

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LOS ALAMOS SCIENTIFIC LABORATORY
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THE USE OF QUARTZ TUBES FOR SAMPLING AND CASTING PLUTONIUM

by

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ABSTRACT

The use of quartz tubing for sampling molten plutonium and for casting rods is described. These procedures offer cheap and easy-to-use methods for obtaining molten samples and for casting long rods with small diameters.

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INTRODUCTION

Plutonium metal rods have been fabricated in the past by pouring molten plutonium into either metal or ceramic molds and machining the product ingot to specifications. Re-usable split copper chill-cast molds[1] have the advantage of producing precision castings, which minimizes subsequent machining; however, they are relatively expensive, thereby increasing the unit cost when only a few rods of a given specification are required. An alternative is to have available several standard molds, but this generally results in undue machining to meet specifications. Cold shuts are also common to the chill-casting techniques unless controlled pouring rates and mold temperatures are used.

In contrast to copper molds, magnesia[2,3] or ceramic-coated graphite molds[4] are inexpensive and can be made by drilling a hole in a rod or block of the mold material. The surface of the product ingot, however, is quite rough and requires considerable machining. Not only are the machining residues increased several fold, but the entire mold must be processed to effect complete recovery of the plutonium. Neither mold

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has been found satisfactory for casting relatively long rods of small (less than 0.075 in.) diameters.

Although quartz is a relatively unreactive material, thermodynamic data[5] indicate that it is attacked by molten plutonium according to the equation

$$SiO_2 + Pu = PuO_2 + Si$$

Experiments verify this reaction, and quartz has been neglected as a container material in plutonium metallurgy. The rate at which this reaction proceeds, however, is a function of the temperature and is relatively slow at temperatures below 1000°C. Therefore, a metallurgical casting process becomes feasible[6] if the temperature and exposure time of the molten plutonium to quartz are limited. With slight modifications the casting process may also be used to remove samples from molten plutonium. Such processes can take advantage of the many desirable physical properties of quartz, namely, a high softening temperature (1500°C), a low coefficient of thermal expansion, an ability to withstand tremendous thermal shock, and a ready availability in a variety of shapes and sizes. In addition, the relatively low thermal conductivity of quartz makes possible the use of chill-casting techniques without the formation of cold shuts even with the quartz at or near room temperature.

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SAMPLING

One of the difficulties encountered in plutonium pyrometallurgical research is that of obtaining samples of a melt during an experiment. The ladle method finds frequent application but is limited due to the size of the apparatus. In a two-phase system a ladle sample of the lower phase is usually contaminated by material from the upper phase. A drainage arrangement may be used to sample the lower of two phases, but the problems encountered are greatly magnified in plutonium metallurgy. The quenching technique, wherein the entire reaction mixture is rapidly cooled and then the solid product sampled, has sources of error. If the rate of change of composition with temperature is very great, then the samples will not be representative of the system at the elevated temperature of the melt. The aforementioned limitations, however, may be circumvented by pressure-differential casting into a quartz tube, similar to a technique used in uranium metallurgy.[6] The procedure is as follows: A porous carbon plug is fitted into a piece of quartz tubing at a predetermined distance from the end. The lower tip of a quartz tube is then lowered into the melt and a vacuum is applied to the upper end of the quartz tube for 1 or 2 seconds to suck the melt up the tube as far as the carbon plug. The quartz tube is then removed from the apparatus and allowed to cool. As the sample cools, a difference in the thermal expansion of the metal and quartz causes the quartz to break away from the sample. Since the quartz is exposed to the molten plutonium for such a short time, only the surface of the sample shows signs of reaction.

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Samples of preselected size may be obtained easily by varying the diameter of the tubing and the position of the porous plug in the tube. In a two-phase system the bottom phase may be sampled by passing an inert gas through the quartz tube as it is lowered into the melt. When the tube has reached the bottom of the container, an uncontaminated sample may be obtained by replacing the pressure with vacuum and following the previously described procedure. Figure 1 shows a typical experimental apparatus in which this sampling procedure is used.

PRESSURE-DIFFERENTIAL CASTING

By varying the position of the porous plug in the aforementioned procedure, it is possible to cast long plutonium or plutonium-alloy rods of 0.05 to 0.25 in. diameter. Voids in the casting due to cold shuts or incomplete filling of the mold are not present in rods made by this method. The quartz residue resulting from these castings may be leached with acid, washed with water, and discarded. Pictured in Fig. 2 are plutonium-alloy rods prepared by pressure-differential casting into quartz.

GRAVITY CASTING

Difficulties are encountered when rods of plutonium or plutonium-rich alloy larger than 0.25 in. in diameter are cast by the pressure-differential method. With pure plutonium the positive coefficient of thermal expansion as the metal cools through the delta-prime and delta

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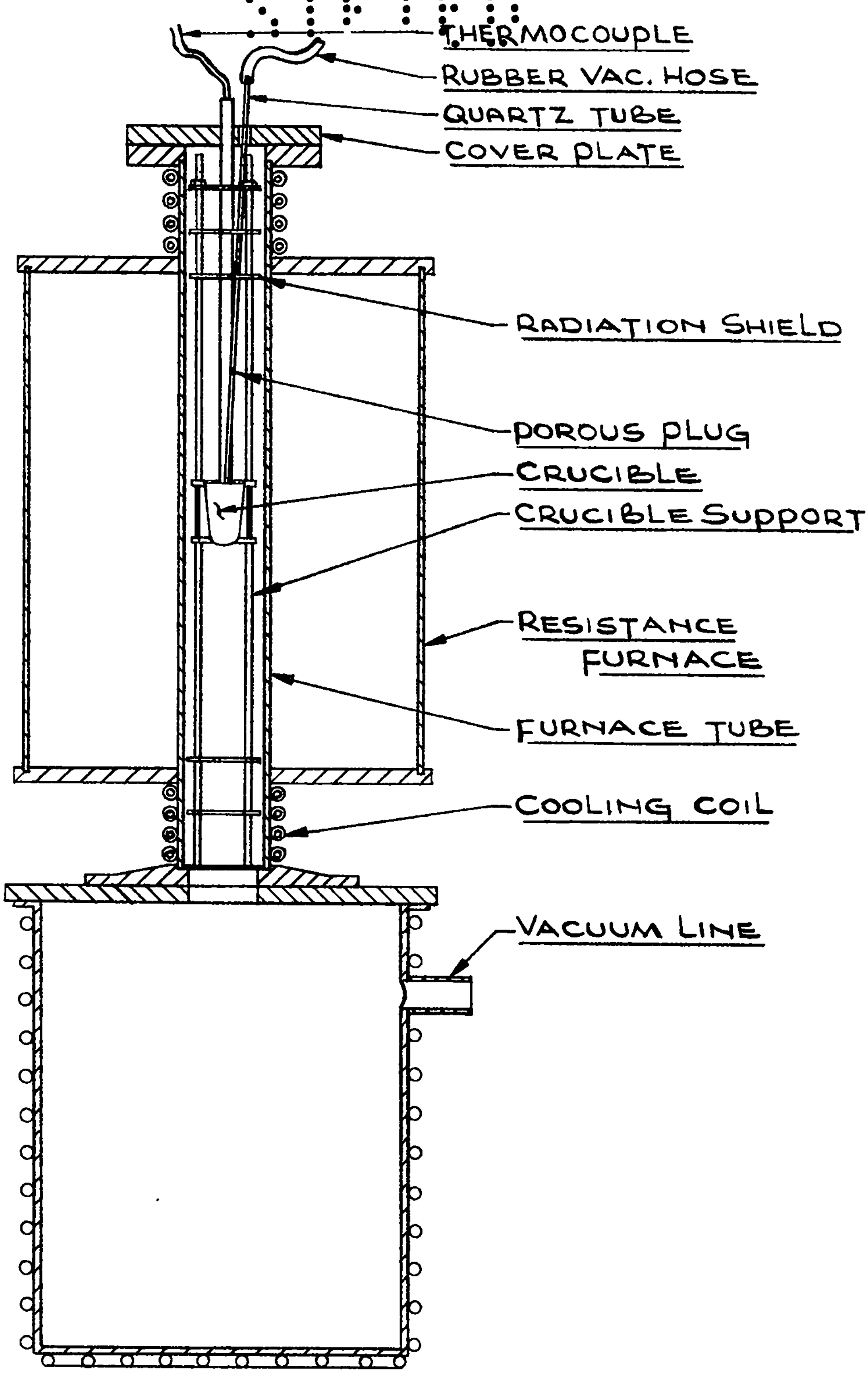

Fig. 1 Sampling schematic.

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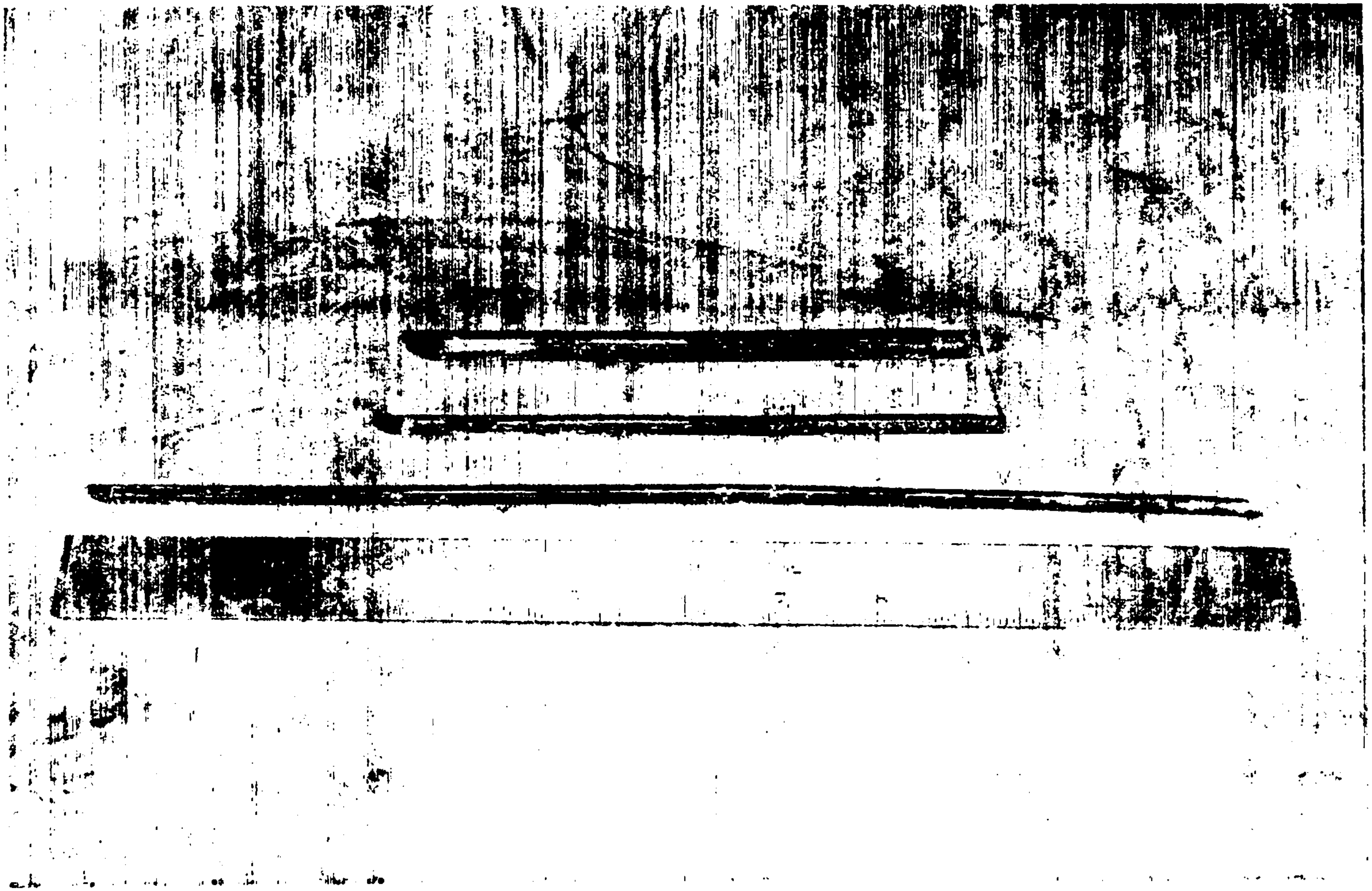

Fig. 2 Rods prepared by pressure-differential casting.

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phases (476° to 319°C)[7] presents a problem. In such castings, a thin ring at the quartz surface solidifies and cools rapidly. As the material cools through the delta-prime and delta phases, it expands, breaking the quartz tube, and molten metal in the center which has not had time to freeze breaks through the thin outer ring. Similarly, certain plutonium alloys, such as the 9.5 atomic per cent iron alloy, expand on solidification and sufficient pressure is exerted to break both the larger size quartz tube and the thin alloy surface which freezes first. Preheating the quartz tubes to a temperature above the delta-phase region prevents this occurrence, but all the advantages inherent in the chill-casting method are then lost.

To cast rods of plutonium or plutonium-rich alloy with a diameter of 0.25 to 0.75 in., the melt is gravity-poured into a quartz tube or tube bundle. Figure 3 is a picture of 0.465 in. diameter, 15 in. long tubes in a three-tube bundle, and Figs. 4a and 4b show castings made in this bundle. The warpage is due to the large decrease in volume as pure plutonium cools through the beta to alpha phase transformation and the surface blemishes are caused by plutonium attack on the tube. Figure 5 is a schematic representation of the casting equipment used. To make such a casting, the feed material is placed in a tantalum or magnesia crucible and heated in a vacuum furnace to approximately 100°C above its melting point. A stopper rod is then pulled, allowing the melt to flow by gravity into the tubes, which are contained in a metal support column. Magnesia sand is packed around the tubes in the

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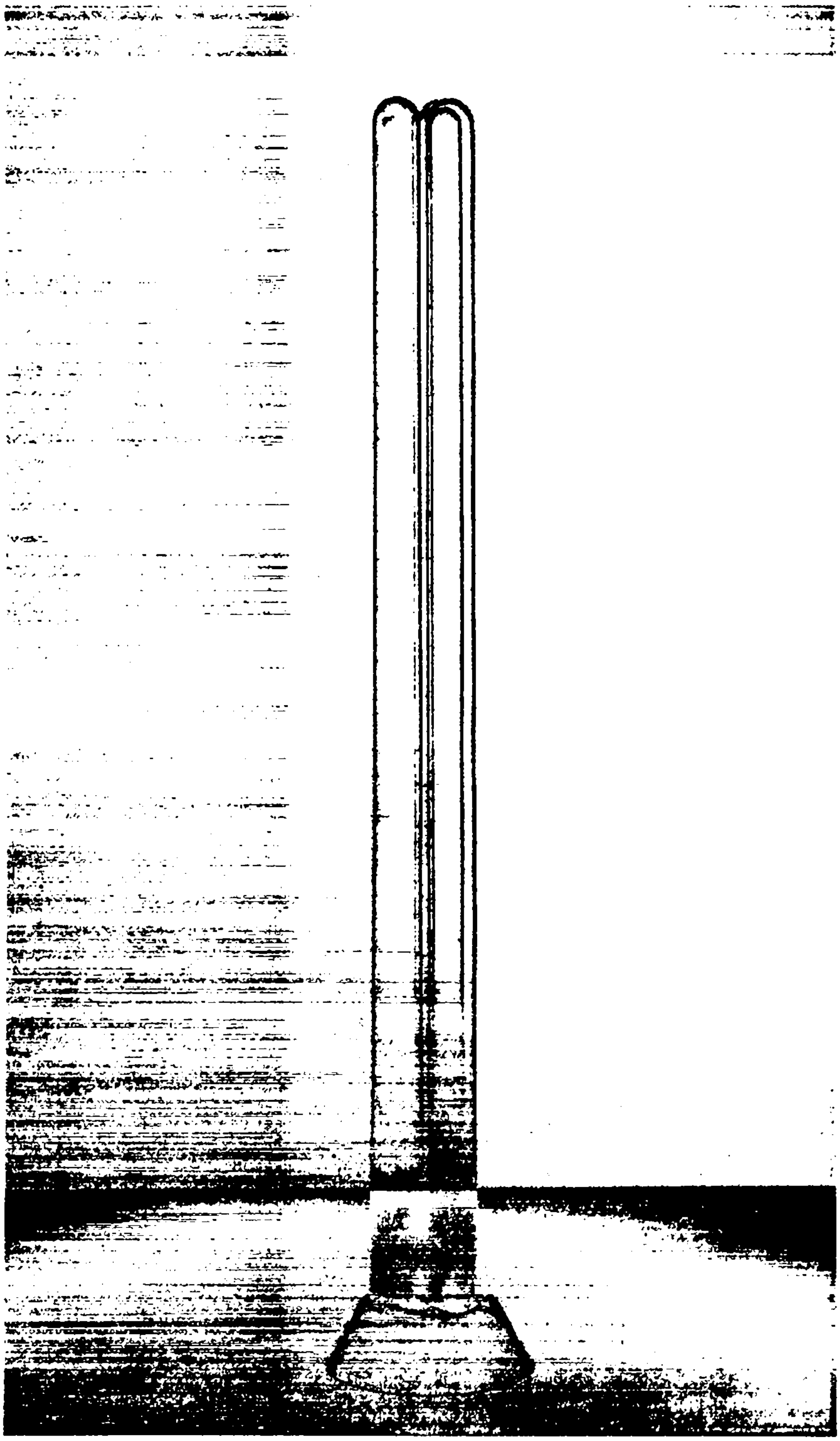

Fig. 3 Quartz mold for gravity casting.

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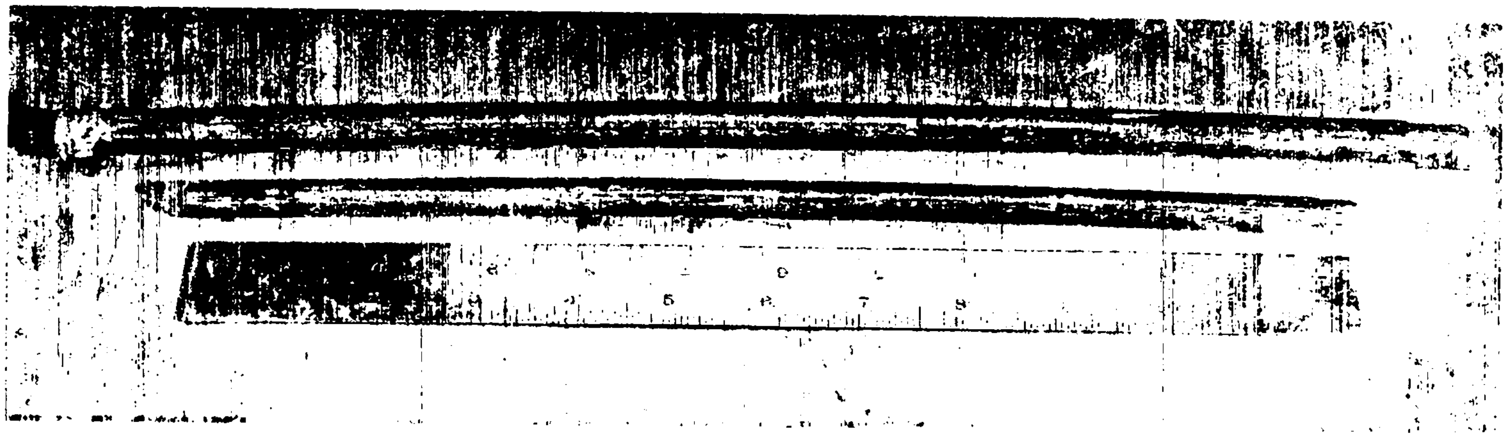

Fig. 4a Plutonium rod bundle gravity cast in quartz tube bundle.

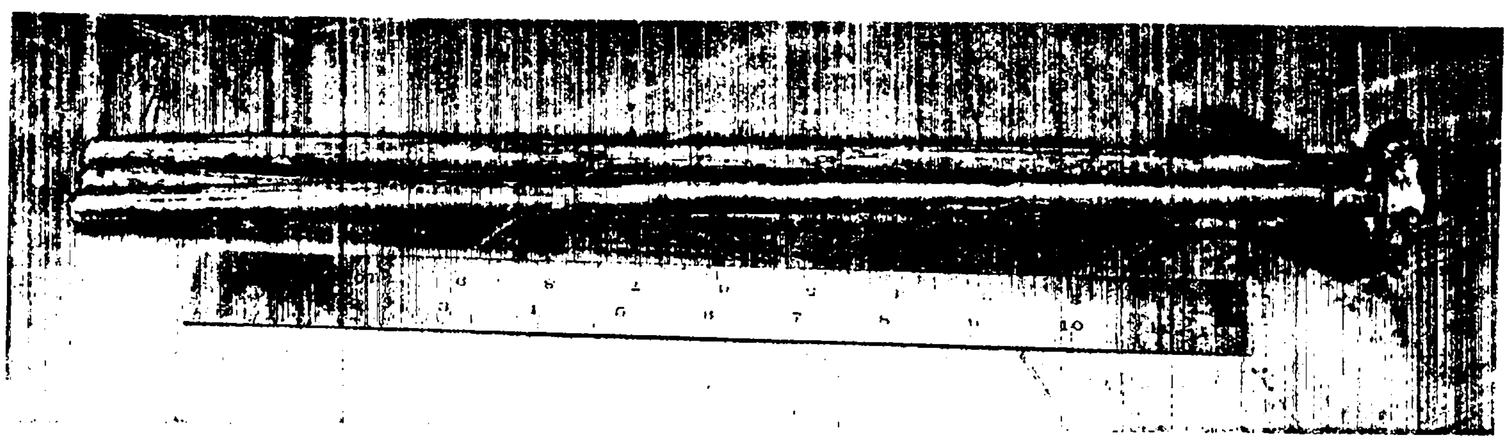

Fig. 4b Plutonium rods gravity cast in quartz tube bundle.

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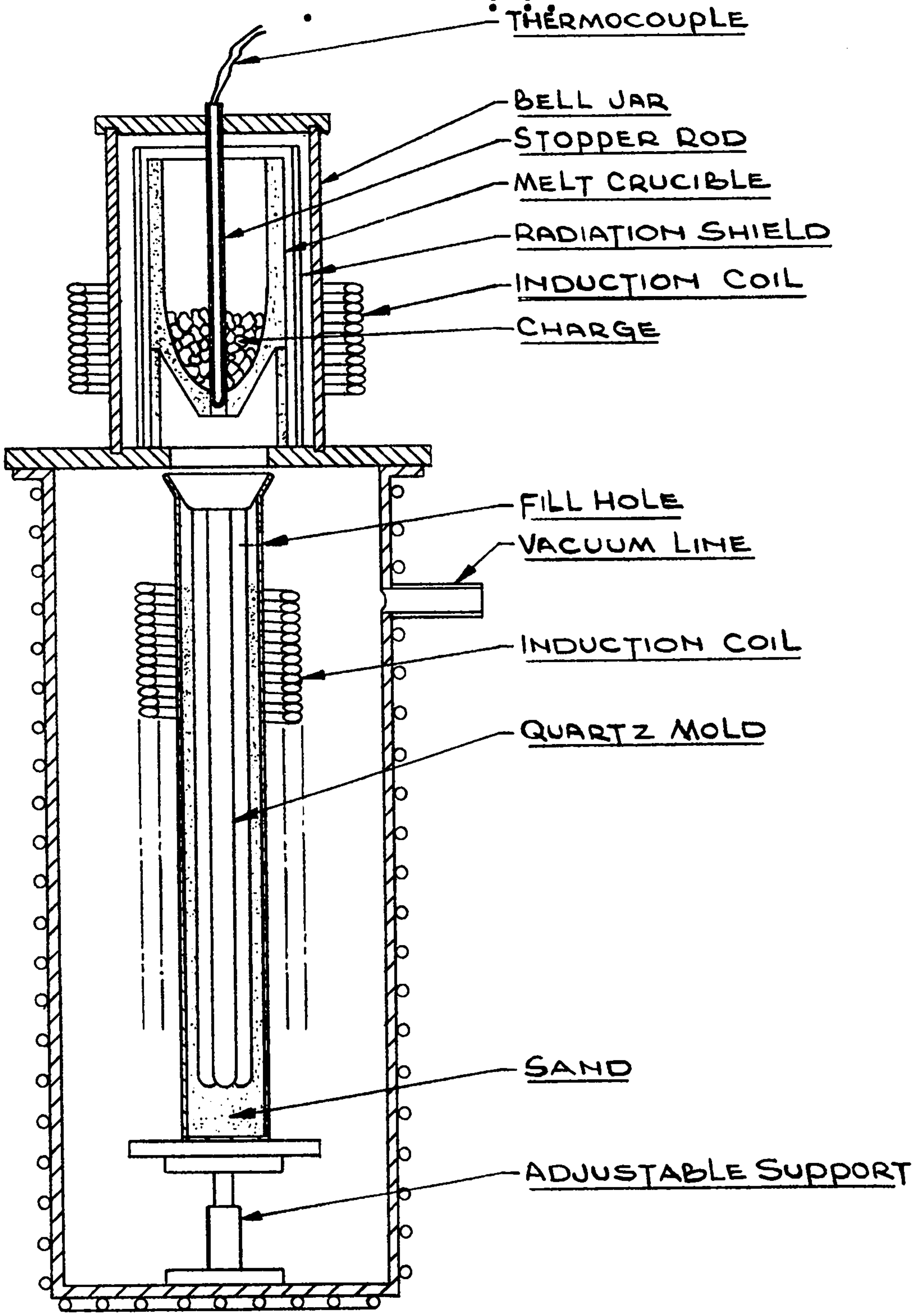

Fig. 5 Schematic of equipment for multiple-rod casting.

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support column to serve as a back-up support during the pour. Depending on the melting point of the material being cast and the amount of super heat used, the tube bundle may or may not be preheated. In any event, sound castings that are free of cold shuts can be made with the tubes at room temperature or slightly above depending on the alloy being poured and the diameter and length of the tubes.

In gravity casting, the positive coefficient of thermal expansion during cooling causes the metal to expand and the quartz to crack; however, the sand packing around the tubes provides sufficient back-up to prevent the liquid melt remaining in the center from breaking through the solidified ring and hence sound rods are obtained. Table I shows the silicon pick-up in a pure plutonium rod cast in this manner.

Table I

SILICON CONTAMINATION IN A ROD CAST IN A QUARTZ TUBE 0.5 IN. IN DIAMETER AND 12 IN. LONG*

Sample	Silicon Concentration, ppm
Feed	190
Rod (skin next to quartz)	630
(0.005 in. below skin)	225
(0.010 in. below skin)	180
(0.015 in. below skin)	190
Center of rod	185

*Melt temperature was 750°C and mold temperature before pour was 275°C.

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Although castings larger than 0.75 in. in diameter can be made in quartz, it is usually more economical to chill-cast them into re-usable metal molds or hot cast into magnesia or ceramic-coated graphite molds.

SUMMARY AND CONCLUSIONS

Although molten plutonium reacts with quartz, the reaction is relatively slow at moderate temperatures. Quartz may therefore be used for removing samples from a melt or as a mold material for casting plutonium or its alloys if the exposure of quartz to the molten metal is minimized. Under these conditions either pure plutonium or plutonium alloys may be cast into rods by pouring the melt into the quartz tubes which are at or near room temperature. Rods may also be cast by a pressure differential method in which molten metal is drawn into a quartz tube by vacuum.

Sampling and casting using quartz has the advantages of being rapid, of permitting the use of a variety of inexpensive molds readily available, and of consistently producing sound castings.

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